

COUPLING SEEPAGE AND RADIONUCLIDE FATE/TRANSPORT IN AND AROUND EMPLACEMENT DRIFTS AT YUCCA MOUNTAIN

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RESEARCH OBJECTIVES

The objectives of this project are to: (1) develop a quantitative model of coupled thermal, hydrological, and chemical (THC) processes potentially leading to brine formation on top of waste packages and/or a drip shield and (2) dynamically integrate such a model into the larger-scale models of processes within and around waste emplacement tunnels, as well as into the smaller-scale waste-package corrosion models.

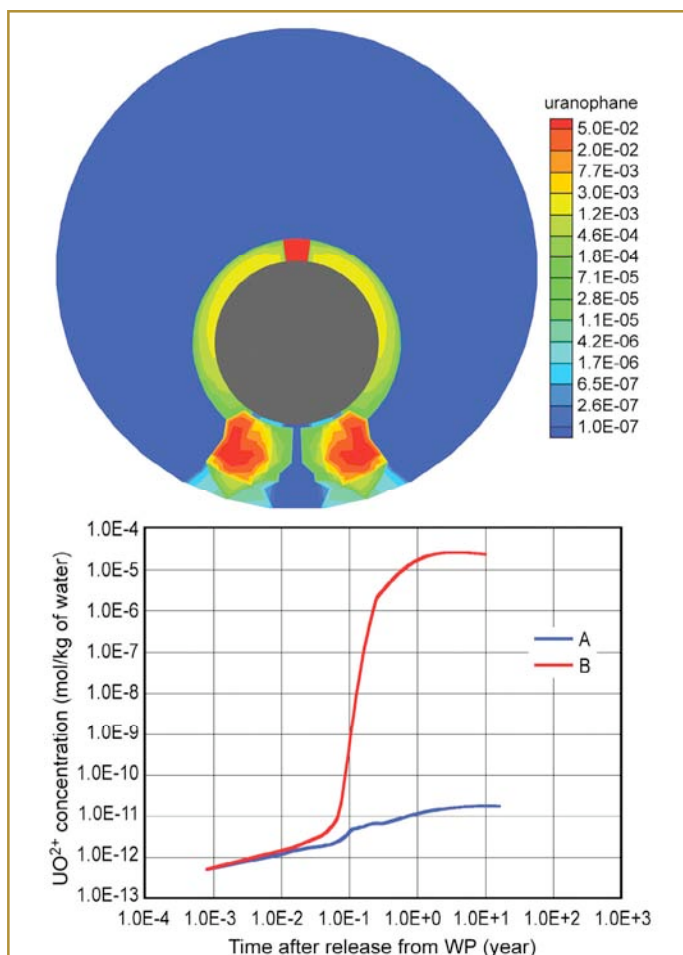


Figure 1. Simulated distribution of uranophane (mol/m³ of medium) precipitated after 25 years of continuous dripping (top) and simulated time evolution of UO_2^{+2} concentration at the base of the drift with (A) and without (B) consideration of uranophane precipitation.

APPROACH

Process models were implemented into TOUGHREACT to allow modeling of (1) evaporative concentration to very high ionic strength, (2) boiling point elevation due to dissolved salts, (3) boiling/evaporation to dryness, and (4) salt deliquescence. An integrated

near-field/in-drift THC simulation was run using a vertical 2-D grid extending from near the ground surface to the groundwater table, and covering a width equal to half the design drift spacing of 81 m. The integrated model was then used to simulate a discrete dripping event within the drift. The model considered the release of radionuclides into seepage water as this water contacts the waste package and flows through the invert. The precipitation of uranophane and Np-uranophane was also considered. These minerals form in the invert from the neutralization of mildly acidic seepage water by clay minerals.

ACCOMPLISHMENTS

The main findings from this modeling effort are as follows: (1) the near-field and in-drift brine chemical evolution is dominated by the precipitation of NaCl, CaSO₄, and CaCO₃; (2) the generation of acid gases at high evaporative concentration yields $P_{HCl} \sim 10^{-7}$ bar at boiling temperatures, with pH staying >5 in condensation areas; (3) the clay minerals in the invert neutralize the pH of seepage water, although this result is sensitive to assumptions regarding the kinetics of reactions with clays; (4) the drift invert may act as a pH buffer that promotes the precipitation of uranophane and impedes further downward migration of radionuclides at elevated concentrations (Figure 1). However, the pH buffer effect is subject to the content and composition of the clay minerals in the invert.

SIGNIFICANCE OF FINDINGS

The model captures some of the processes involved in salt formation and radionuclide transport, and can be further applied to capturing the details of radionuclide transport between the waste form and the rock through the invert.

RELATED PUBLICATIONS

- Zhang, G., N. Spycher, E. Sonnenthal, and C. Steefel, Implementation of a Pitzer activity model into TOUGHREACT for modeling concentrated solutions. Proceedings, TOUGH2 Symposium, Lawrence Berkeley National Laboratory, Berkeley, CA, May 15–17, 2006.
- Zhang, G., N. Spycher, T. Xu, E. Sonnenthal, and C. Steefel, Reactive geochemical transport modeling of concentrated aqueous solutions: Supplement to the TOUGHREACT Users's Guide for the Pitzer ion-interaction model. LBNL-62718, Lawrence Berkeley National Laboratory, Berkeley, CA, 2007.

ACKNOWLEDGMENTS

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